Global and Local Measures of the Intrinsic Josephson Coupling in $Tl_2Ba_2CuO_6$

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One leading candidate theory of the high-temperature superconductors in the copper oxide systems is the Inter-Layer Tunneling (ILT) mechanism[1]. In this model superconductivity is created by tunneling of electron pairs between the copper oxdide planes-contrasting with other models in which superconductivty first arises by electron pairing within each plane. The ILT model predicts that the superconducting condensation energy is approximately equal to the gain in kinetic energy of the electron pairs due to tunneling. Both these energies can be determined independently[2-4], providing a quantitative test of the model. The gain in kinetic energy of the electron pairs is related to the interlayer plasma energy, ω_J, of electron pair oscillations, which can be measured using infrared spectroscopy. Direct imaging of magnetic flux vortices also provides a test[5], which is performed here on the same samples. In the high-temperature superconductor $Tl_2Ba_2CuO_6$, both the sample averaging optical probe and the local vortex imaging give a consistent value of $\omega_J = 28$ cm^{-1} which, when combined with the condensation energy produces a discrepancy of at least an order of magnitude with deductions based on the ILT model.

In the ILT model the normal state is different in nature from the traditional Landau Fermi liquid. As a result coherent transport of single charge carriers between the planes is strongly inhibited in the normal state. In the superconducting phase tunneling of pairs is possible, and the superconducting condensation energy (E_{cond}) in the ILT model is precisely the gain in kinetic energy (E_J) due to the tunneling of those pairs: $E_J = \eta E_{cond}$. The number η is of order 1 when ILT is the only active pairing mechanism[3]. With conventional mechanisms, although usually $\eta \ll 1$, there is no prediction for η that is free from materials parameters. A crucial point in this discussion is, that both E_{cond} and E_J are experimentally accessible quantities, thus allowing the experimental verification of the ILT hypothesis. E_{cond} can be measured from the specific heat[6]. E_J can be determined by measuring the interlayer (Josephson) plasma frequency[7].

For this work, we used two kinds of samples: single crystals and epitaxial thin films of $Tl_2Ba_2CuO_6$. The crystals have a transition temperature of 82K and transition width $(10\% - 90\%)$ of 13 K, as determined by bulk SQUID susceptibility. Using 4-circle X-ray diffraction we verified that the material belongs to the tetragonal $I4/mmm$ space group, with (for the crystals) lattice parameters a=b=3.867Å , and c=23.223Å . The films have T_c=80K as determined by DC resistivity, and c=23.14 Å . Both types of samples have relatively large physical dimensions perpendicular to the c-axis, corresponding to the con-
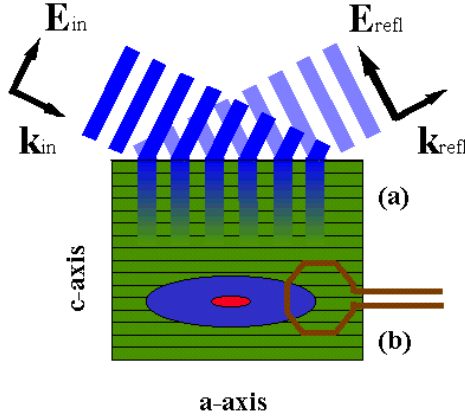
Figure 1: *Schematic of the experimental techniques: a) Grazing incidence reflectivity. The p-polarized light incident at a grazing angle sets up a periodic electric field pattern, which is polarized perpendicular to the sample surface, and decays exponentially inside the solid. b) Scanning SQUID microscopy. The octagonal pickup loop detects the magnetic flux perpendicular to the a-c face.*

ducting copper oxide planes (50 mm^2 for the thin films, and 1 mm^2 for the crystals). They have small dimensions along the c-axis (1 μm for the thin films, and 50 μm for the crystals).

To determine the plasma resonance we measure the reflection coefficient of infrared radiation incident on the ab-plane at a large angle (80°) with the surface normal[8]. A sketch of the experiment is presented in Fig. 1. In the case of the single crystals the reflected light drops below our detection limit if the wavelength exceeds 0.2 mm (i.e. for $\omega/2\pi c < 50 cm^{-1}$) due to diffraction. Using thin films we were able to extend this range to 20 cm^{-1}. The electric field vector of the radiation is chosen parallel to the plane of reflection, resulting in a large component perpendicular to the CuO$_2$ planes. This geometry allows absorption of the light by lattice vibrations and plasma-oscillations polarised perpendicular to the planes.

In Fig. 2a we present the single crystal and thin film reflectivity for $\omega/2\pi$c respectively above and below 150 cm^{-1}. All prominent absorption lines for frequencies above 50 cm^{-1} correspond to infrared active lattice vibrations, which show no strong temperature dependence. In the 4K spectrum we observe a clear absorption at 27.8 cm^{-1}. This resonance exhibits a strong red shift upon raising the temperature, as displayed in Fig. 2b. Above 70 K it has shifted outside our spectral window. In Fig. 2c we also present the temperature dependence of $\omega(T)^2/\omega(4K)^2$ of the resonance position. This temperature dependence extrapolates to zero at T$_c$, which indicates, that it is a plasma resonance of the paired charge carriers. We therefore attribute this absorption to a Josephson plasmon, a collective oscillation of the paired charge carriers perpendicular to the coupled superconducting planes[7]. For a purely electronic system the supercurrent density along the c-axis determines the Josephson resonance frequency, c/λ_c. Because in the present case the Josephson plasma resonance is located at a frequency below the infrared active
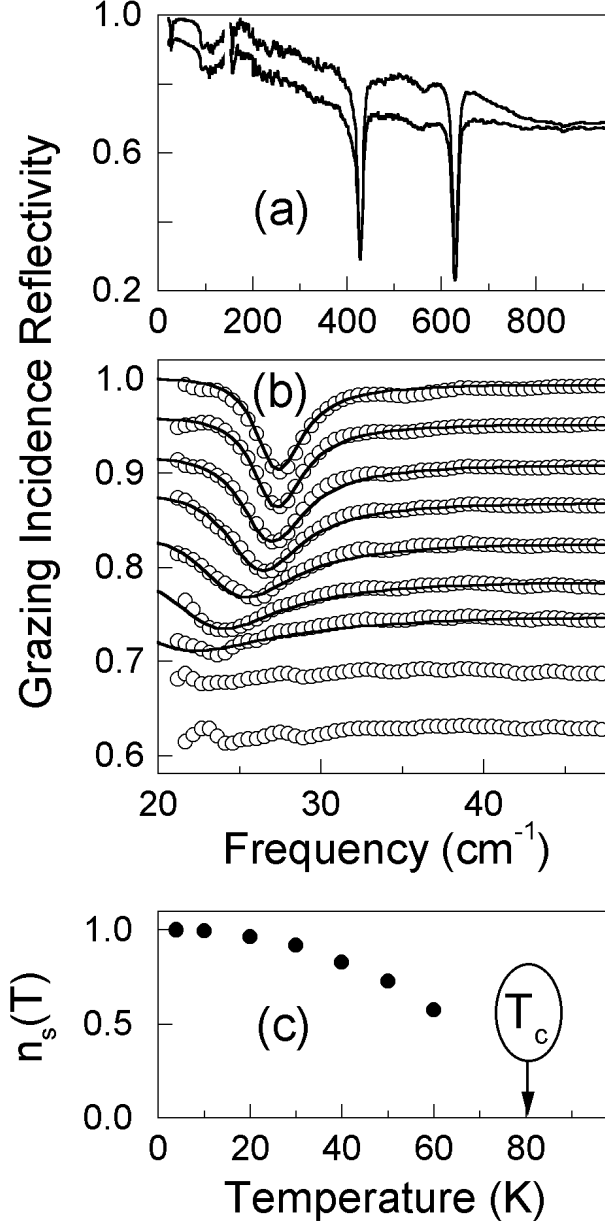
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Figure 2: a) P-polarized reflectivity at 80° angle of incidence of $Tl_2Ba_2CuO_6$ at 4 K (uppercurve) and 100 K (lower curve). Frequencies above (below) 150 cm^{-1} correspond to single crystal (thin film) data. b) Thin film spectra on an expanded frequency scale. From top to bottom: 4K, 10 K, 20 K, 30 K, 40 K, 50 K, 60 K, 75 K, nd 90 K. The curves have been given incremental 3 percent vertical offsets for clarity. The solid curves correspond to calculations as described in the text. c) Temperature dependence of $n_s(T) = \omega_J(T)^2/\omega_J(4K)^2$, demonstrating that the resonance frequency converges to zero at T_c.
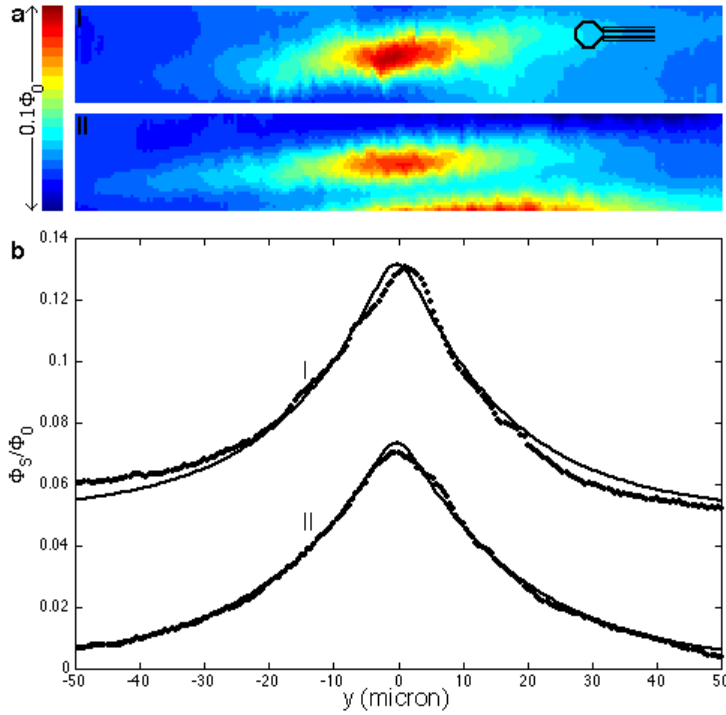
Figure 3: a) Images of the magnetic field perpendicular to an a-c face of a $Tl_2Ba_2CuO_6$ single crystal in two different locations, showing two different interlayer Josephson vortices. The dashed lines indicate the longitudinal cross-sections. Inset: sketch of the 4 mm octagonal pickup loop. b) The flux through the SQUID pickup loop along the longitudinal cross-sections. The solid curves are fits which determine the c-axis penetration depths of these two vortices to be (I) $\lambda_c = 17 \pm 4$ mm and (II) $\lambda_c = 1 \pm 1$ mm. The straight lines indicate the extent of λ_c for each vortex.

lattice vibrations, the corresponding dynamical electric field is screened by the ions and the lattice vibrations, characterised by a dielectric constant ecs. As a result we observe the Josephson resonance at a reduced frequency $\omega_J = \epsilon_{cs}^{-1/2} c/\lambda_c$. We performed a full optical analysis of these spectra in the spectral range from 20 to 6000 cm^{-1} using Fresnel's equations for oblique angle of incidence reflection of anisotropic optical media. This way we were able to extract the dielectric function ecs from our data. For frequencies below 40 cm^{-1}, $\epsilon_{cs} = 11.3 \pm 0.5$. We therefore obtain $\lambda_c(4K) = 17.0 \pm 0.3 \mu m$.

An independent experimental measure of the interlayer coupling is provided by a direct measurement of λ_c. Here we employ the fact that vortices which are oriented parallel to the planes, called interlayer Josephson vortices, have a characteristic size λ_c along the planes and λ_a perpendicular to the planes[9]. In order to determine λ_c directly, we used a scanning Superconducting QUantum Interference Device (SQUID) microscope10 to map the magnetic fields perpendicular to an a-c face at 4 K (Fig. 1b) [5]. The crystal was cooled in a magnetically shielded cryostat with a residual magnetic field of a few milligauss, resulting in the presence of a few isolated trapped vortices (Fig. 3a). The jitter apparent in this image is due to the mechanical scanning mechanism used in our SQUID microscope. With an L = 4 mm octagonal pickup loop, the vortices were resolution-limited along the c direction ($\lambda_a \ll L$), but not along the a direction ($\lambda_c \ll L$).

Fitting the longitudinal cross-sections (Fig. 3b) to the functional form for the magnetic fields of an interlayer Josephson vortex[9] convoluted with the shape of the pickup loop[5,10], gave the results $\lambda_c = 17 \pm 4$ mm and $\lambda_c = 19 \pm 1$ mm for these two vortices. The statistical error bars were determined using a criterion of doubling of the variance from the least-squares value, but systematic errors from the background and the shape of the pickup loop, and the effect of the surface on the shape of the vortex, which may be as large as 30vortices in three pieces cut from a large single crystal, which was part of the mosaic used to make the measurements in Fig. 1a. The vortices in all three pieces confirm the plasma resonance frequency of 28cm^{-1}.

We are now ready to determine E$_J$ using the expression[7] $\epsilon_{cs}\hbar^2\omega_J^2 = 4\pi da^{-2}e^{*2}E_J$, where d is the distance between planes (11.6$\overset{\circ}{A}$), a is the cell parameter (3.87$\overset{\circ}{A}$), $\hbar$ is the Planck constant, and $e^* = 2e$ is the charge of the pairs. The result is $E_J = 0.24\mu eV$ per formula unit. For Tl$_2$Ba$_2$CuO$_6$ the measured value of E$_{cond}$ is 100 $\pm 20\mu eV$ per formula unit[6]. Hence $\eta = E_J/E_{cond} = 0.0024 \pm 0.0005$, which is clearly at variance with the notion that condensation in the high-T$_c$ superconductors is due to the gain in kinetic energy of the pairs (E$_J$) in the superconducting state.

Our observation of the same value of ω_J with a local probe (scanning SQUID) and a sample averaging probe (grazing reflectivity) demonstrates that we measure the intrinsic Josephson coupling, rather than a coupling determined by isolated metallurgical defects. The body of data presented in this paper provides strong support for the interpretation of both the Josephson plasma resonance and the interlayer Josephson vortices as intrinsic properties of Tl$_2$Ba$_2$CuO$_6$. One of the key predictions of the ILT model, that η=E$_J$/E$_{cond}$ is of order 1, is far outside the range of our experiments, which give η=0.0024.

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